

BRIX, and identified by Attorney Docket No. NHL-SCT-18 US.

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A This application is also related to Application Serial No. 09/758,946, filed concurrently herewith on January 11, 2001, having the title ALKALI-FREE ALUMINOBOROSILICATE GLASS, AND USES THEREOF, naming as inventors Dr. Ulrich PEUCHERT and Dr. Peter BRIX, and identified by Attorney Docket No. NHL-SCT-20 US.

09/758,952 - 043001  
This application is further related to Application Serial No. 09/758,903, filed concurrently herewith on January 11, 2001, having the title ALKALI-FREE ALUMINOBOROSILICATE GLASS, AND USES THEREOF, naming as inventors Dr. Ulrich PEUCHERT and Dr. Peter BRIX, and identified by Attorney Docket No. NHL-SCT-21 US.--

The paragraph beginning at page 5, line 20, has been amended as follows:

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A --The glasses described in Japanese Patent Application No. 9-12333 A for hard disks, are comparatively low in  $Al_2O_3$  or  $B_2O_3$ , the latter merely being optional. The glasses have high alkaline earth metal oxide contents and have high thermal expansion, which makes them unsuitable for use in LCD or PV technology.--

The paragraphs beginning at page 7, line 4, and ending on page 9, line 7, have been amended as follows:

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A --The invention teaches that this object can be accomplished by aluminoborosilicate glasses having a coefficient of thermal expansion  $\alpha_{20/300}$  of between  $2.8 \times 10^{-6}/K$  and  $3.8 \times 10^{-6}/K$ , which has the following composition (in % by weight, based on oxide):

silicon dioxide ( $\text{SiO}_2$ ) - from somewhat more than 58% to 65% (> 58% - 65%); boric oxide ( $\text{B}_2\text{O}_3$ ) - from somewhat more than 6% to 10.5% (> 6% - 10.5%); aluminum oxide ( $\text{Al}_2\text{O}_3$ ) - from somewhat more than 14% to 25% (> 14% - 25%); magnesium oxide ( $\text{MgO}$ ) from 0% to somewhat less than 3% (0 - < 3%); calcium oxide ( $\text{CaO}$ ) - from 0% to 9% (0% - 9%); strontium oxide ( $\text{SrO}$ ) - from 0.1% to 1.5% (0.1% - 1.5%); barium oxide ( $\text{BaO}$ ) - from somewhat more than 5% to 8.5% (> 5% - 8.5%); with strontium oxide ( $\text{SrO}$ ) + barium oxide ( $\text{BaO}$ ) - equal to or somewhat less than 8.6% ( $\leq 8.6\%$ ); and with magnesium oxide ( $\text{MgO}$ ) + calcium oxide ( $\text{CaO}$ ) + strontium oxide ( $\text{SrO}$ ) + barium oxide ( $\text{BaO}$ ) - from 8% to 18% (8% - 18%); and zinc oxide ( $\text{ZnO}$ ) - from 0% to somewhat less than 2% (0% - < 2%).

The invention also teaches an alkali-free aluminoborosilicate glass having a coefficient of thermal expansion  $\alpha_{20/300}$  of between  $2.8 \times 10^{-6}/\text{K}$  and  $3.6 \times 10^{-6}/\text{K}$ , which has the following composition (in % by weight, based on oxide): silicon dioxide ( $\text{SiO}_2$ ) - from somewhat more than 58% to 64.5% (> 58% - 64.5%); boric oxide ( $\text{B}_2\text{O}_3$ ) - from somewhat more than 6% to 10.5% (> 6% - 10.5%); aluminum oxide ( $\text{Al}_2\text{O}_3$ ) - from 20.5% to 24% (20.5% - 24%); magnesium oxide ( $\text{MgO}$ ) - from 0% to somewhat less than 3% (0% - < 3%); calcium oxide ( $\text{CaO}$ ) - from 2.5% to somewhat less than 8% (2.5% - < 8%); strontium oxide ( $\text{SrO}$ ) - from 0.1% to 3.5% (0.1% - 3.5%); barium oxide ( $\text{BaO}$ ) - from somewhat more than 5% to 7.5% (> 5% - 7.5%); with strontium oxide ( $\text{SrO}$ ) +

barium oxide (BaO) being equal to or less than 8.6% ( $\leq 8.6\%$ ); and with magnesium oxide (MgO) + calcium oxide (CaO) + strontium oxide (SrO) + barium oxide (BaO) in the range of from 8% to 18% (8% - 18%); and zinc oxide (ZnO) - from 0% to somewhat less than 2% (0% -  $< 2\%$ ).

The glass contains between  $> 58$  and 65% by weight of  $\text{SiO}_2$ . At lower contents, the chemical resistance is impaired, while at higher levels, the thermal expansion is too low and the crystallization tendency of the glass increases. Preference is given to a maximum content of 64.5% by weight.

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The glass contains relatively high levels of  $(\text{Al}_2\text{O}_3)$ , i.e.  $> 14 - 25\%$  by weight of  $(\text{Al}_2\text{O}_3)$ , preferably at least 18% by weight, particularly preferably  $> 18\%$  by weight. These relatively high  $\text{Al}_2\text{O}_3$  levels are favourable to the crystallization stability of the glass and have a positive effect on its heat resistance without excessively increasing the processing temperature. Particular preference is given to a content of at least 20.5% by weight, most preferably of at least 21.5% by weight, of  $(\text{Al}_2\text{O}_3)$ . Preference is given to a maximum  $\text{Al}_2\text{O}_3$  content of 24% by weight.

The  $\text{B}_2\text{O}_3$  content is  $> 6 - 10.5\%$  by weight. The  $\text{B}_2\text{O}_3$  content is restricted to the maximum content specified in order to achieve a high glass transition temperature  $T_g$ . Higher contents would also impair the chemical resistance to hydrochloric acid solutions. (Preference is given to a maximum  $\text{B}_2\text{O}_3$  content of 11%

by weight). The minimum  $B_2O_3$  content specified serves to ensure that the glass has good meltability and good crystallization stability. Preference is given to a minimum content of  $> 8\%$  by weight of  $B_2O_3$ .

The network-forming components ( $Al_2O_3$ ) and  $B_2O_3$  are preferably present at mutually dependent minimum levels, ensuring a sufficient content of the network formers  $SiO_2$ , ( $Al_2O_3$ ) and  $B_2O_3$ . For example, in the case of a  $B_2O_3$  content of  $> 6 - 10.5\%$  by weight, the minimum  $Al_2O_3$  content is preferably  $> 18\%$  by weight, and in the case of an ( $Al_2O_3$ ) content of  $> 14 - 25\%$  by weight, the minimum  $B_2O_3$  content is preferably  $> 8\%$  by weight. Preferably, in particular in order to achieve low thermal expansion coefficients of up to  $3.6 \times 10^{-6}/K$ , the sum of  $SiO_2$ ,  $B_2O_3$  and  $Al_2O_3$  is at least  $85\%$  by weight.--

The paragraph beginning at page 10, line 6, has been amended as follows:

--On the other hand, the maximum  $SrO$  content can be up to  $3.5\%$  by weight in the case of high- $Al_2O_3$  (in particular  $\geq 20.5\%$  by weight) and relatively  $CaO$ -rich (in particular  $\geq 2.5\%$  by weight) glasses. The higher  $SrO$  content has the positive effect of counteracting the slight increase in crystallization tendency found in relatively  $CaO$ -rich glasses having relatively high  $Al_2O_3$  contents.--

The paragraph beginning at page 11, line 10, has been

amended as follows:

AS  
--In addition to the low-SrO glass of the main claim, a glass having the desired requirement profile and a coefficient of thermal expansion  $\alpha_{20/300}$  of between  $2.8 \times 10^{-6}/K$  and  $3.6 \times 10^{-6}/K$  is also described by the following composition (in % by weight, based on oxide):  $SiO_2 > 58 - 64.5$ ,  $B_2O_3 > 6 - 10.5$ ,  $Al_2O_3 > 20.5 - 24$ ,  $MgO 0 - < 3$ ,  $CaO 2.5 - < 8$ ,  $SrO 0.1 - 3.5$  and  $BaO > 5 - 7.5$ , with  $SrO + BaO \leq 8.6$  and with  $MgO + CaO + SrO + BaO 8 - 18$ ;  $ZnO 0 - < 2$ .--

09758952-043001  
The paragraph beginning at page 12, line 5, has been amended as follows:

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--The glass may contain conventional refining agents in the usual amounts: it may thus contain up to 1.5% by weight of  $As_2O_3$ ,  $Sb_2O_3$ ,  $SnO_2$ ,  $CeO_2$ ,  $Cl^-$  (for example in the form of  $BaCl_2$ ),  $F^-$  (for example in the form of  $CaF_2$ ) and/or  $SO_4^{2-}$  (for example in the form of  $BaSO_4$ ). The sum of the refining agents should, however, not exceed 1.5% by weight. If the refining agents  $As_2O_3$  and  $Sb_2O_3$  are omitted, the glass can be processed not only using a variety of drawing methods, but also by the float method.--

The portion of the table located on page 16 has been amended as follows:

|                                   | 7     | 8     | 9     | 10    | 11    | 12   |
|-----------------------------------|-------|-------|-------|-------|-------|------|
| SiO <sub>2</sub>                  | 58.2  | 58.1  | 60.5  | 61.5  | 62.0  | 61.0 |
| B <sub>2</sub> O <sub>3</sub>     | 7.6   | 7.6   | 9.5   | 9.6   | 9.5   | 6.2  |
| Al <sub>2</sub> O <sub>3</sub>    | 21.4  | 21.5  | 18.2  | 17.1  | 16.5  | 18.5 |
| MgO                               | 2.8   | 2.8   | 1.9   | 1.9   | 2.7   | 1.0  |
| CaO                               | 2.5   | 2.5   | 2.6   | 2.6   | 1.3   | 6.0  |
| SrO                               | 2.0   | 1.0   | 1.0   | 0.5   | 0.7   | 1.0  |
| BaO                               | 5.2   | 5.2   | 6.0   | 6.5   | 7.0   | 5.5  |
| ZnO                               | —     | 1.0   | —     | —     | —     | 0.5  |
| $\alpha_{20/300}$ [ $10^{-6}/K$ ] | 3.18  | 3.09  | 3.04  | 3.04  | 3.02  | 3.46 |
| $\rho$ [g/cm <sup>3</sup> ]       | 2.51  | 5.52  | 2.46  | 2.44  | 2.48  | 2.53 |
| T <sub>g</sub> [°C]               | 747   | 742   | 727   | 723   | 715   | 740  |
| T4 [°C]                           | 1303  | 1305  | 1320  | 1325  | 1309  | 1315 |
| T2 [°C]                           | 1655  | 1660  | 1671  | 1678  | 1681  | n.m. |
| n <sub>d</sub>                    | 1.522 | 1.522 | 1.514 | 1.512 | 1.510 | n.m. |
| HCl [mg/cm <sup>2</sup> ]         | n.m.  | n.m.  | n.m.  | n.m.  | n.m.  | n.m. |
| BHF [mg/cm <sup>2</sup> ]         | 0.65  | 0.64  | 0.53  | 0.50  | 0.52  | n.m. |

The portion of the table located on page 17 has been amended as follows:

|                                   | 13    | 14    | 15    | 16    |
|-----------------------------------|-------|-------|-------|-------|
| SiO <sub>2</sub>                  | 59.9  | 58.9  | 59.9  | 59.7  |
| B <sub>2</sub> O <sub>3</sub>     | 8.5   | 8.5   | 6.5   | 8.0   |
| Al <sub>2</sub> O <sub>3</sub>    | 15.5  | 16.5  | 18.5  | 20.5  |
| MgO                               | 2.0   | 0.6   | 2.8   | 1.6   |
| CaO                               | 7.2   | 8.2   | 6.0   | 2.5   |
| SrO                               | 1.0   | 0.5   | 0.5   | 2.0   |
| BaO                               | 5.1   | 5.5   | 5.5   | 5.1   |
| ZnO                               | 0.5   | 1.0   | —     | —     |
| $\alpha_{20/300}$ [ $10^{-6}/K$ ] | 3.74  | 3.75  | 3.57  | 3.03  |
| $\rho$ [g/cm <sup>3</sup> ]       | 2.52  | 2.53  | 2.53  | 2.495 |
| T <sub>g</sub> [°C]               | 706   | 708   | 737   | 740   |
| T4 [°C]                           | 1264  | 1266  | 1291  | 1324  |
| T2 [°C]                           | 1623  | 1624  | 1646  | 1708  |
| n <sub>d</sub>                    | 1.524 | 1.526 | 1.526 | 1.517 |
| HCl [mg/cm <sup>2</sup> ]         | 0.38  | 0.37  | 0.27  | 0.99  |
| BHF [mg/cm <sup>2</sup> ]         | 0.53  | 0.51  | 0.58  | 0.59  |

The paragraph beginning at page 18, line 26, has been amended as follows:

--The glasses have high thermal shock resistance and good devitrification stability. The glasses can be produced as flat glasses by the various drawing methods, for example microsheet

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down-draw, up-draw or overflow fusion methods, and, in a preferred embodiment, if they are free from  $\text{As}_2\text{O}_3$  and  $\text{Sb}_2\text{O}_3$ , also by the float process.--

The paragraph beginning at page 21, line 18, has been amended as follows:

A<sup>10</sup>  
--Another feature of the invention resides broadly in an aluminoborosilicate glass, characterized in that it comprises at least 18% by weight, preferably more than 18% by weight, of  $\text{Al}_2\text{O}_3$ .--

TOE40-2568260  
The paragraph beginning at page 21, line 28, and ending on page 22, line 3, has been amended as follows:

A<sup>11</sup>  
--A further feature of the invention resides broadly in an alkali-free aluminoborosilicate glass having a coefficient of thermal expansion  $\alpha_{20/300}$  of between  $2.8 \times 10^{-6}/\text{K}$  and  $3.6 \times 10^{-6}/\text{K}$ , which has the following composition (in % by weight, based on oxide):  $\text{SiO}_2 > 58 - 64.5$ ;  $\text{B}_2\text{O}_3 > 6 - 10.5$ ;  $\text{Al}_2\text{O}_3 20.5 - 24$ ;  $\text{MgO } 0 - < 3$ ;  $\text{CaO } 2.5 - < 8$ ;  $\text{SrO } 0.1 - 3.5$ ;  $\text{BaO } > 5 - 7.5$ ; with  $\text{SrO} + \text{BaO} \leq 8.6$ ; with  $\text{MgO} + \text{CaO} + \text{SrO} + \text{BaO } 8 - 18$ ;  $\text{ZnO } 0 - < 2$ .--

The paragraph beginning at page 22, line 13, has been amended as follows:

A<sup>12</sup>  
--A further feature of the invention resides broadly in an aluminoborosilicate glass, characterized in that it additionally comprises:  $\text{ZrO}_2 0 - 2$ ;  $\text{TiO}_2 0 - 2$ ; with  $\text{ZrO}_2 + \text{TiO}_2 0 - 2$ ;  $\text{As}_2\text{O}_3 0 - 1.5$ ;  $\text{Sb}_2\text{O}_3 0 - 1.5$ ;  $\text{SnO}_2 0 - 1.5$ ;  $\text{CeO}_2 0 - 1.5$ ;  $\text{Cl}^- 0 - 1.5$ ;  $\text{F}^- 0$



A<sup>12</sup> - 1.5;  $\text{SO}_4^{2-}$  0 - 1.5; with  $\text{As}_2\text{O}_3$  +  $\text{Sb}_2\text{O}_3$  +  $\text{SnO}_2$  +  $\text{CeO}_2$  +  $\text{Cl}^-$  +  $\text{F}^-$  +  
 $\text{SO}_4^{2-} \leq 1.5$ .--

The paragraphs beginning at page 23, line 25, and ending on page 24, line 26, have been amended as follows:

A<sup>13</sup> 09758952-043001  
--The corresponding foreign and international patent publication applications, namely, Federal Republic of Germany Patent Application No. 100 00 836.4-45, filed on January 12, 2000, having inventors Dr. Ulrich PEUCHERT and Dr. Peter BRIX, as well as their published equivalents, and other equivalents or corresponding applications, if any, in corresponding cases in the Federal Republic of Germany and elsewhere, and the references cited in any of the documents cited herein, are hereby incorporated by reference as if set forth in their entirety herein, are hereby incorporated by reference as if set forth in their entirety herein.

The corresponding foreign and international patent publication applications, namely, Federal Republic of Germany Patent Application No. 100 00 839.9-45, filed on January 12, 2000, having inventors Dr. Ulrich PEUCHERT and Dr. Peter BRIX, as well as their published equivalents, and other equivalents or corresponding applications, if any, in corresponding cases in the Federal Republic of Germany and elsewhere, and the references cited in any of the documents cited herein, are hereby incorporated by reference as if set forth in their entirety